IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:

Furman et al.

Serial No.: 09/726,769

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For: Methods of Functionalizing and Functionalized Metal Oxide Particles and Mechanically Strong and Transparent or Translucent Composites Made

Using Zirconium Oxide Nanoparticles

MARKED UP CHANGES FILED WITH RESPONSE TO FIRST OFFICE ACTION

Assistant Commissioner for Patents Washington, D.C. 20231

AMENDMENTS

In the Specification

1. Page 13, paragraph beginning at line 3:

The silyl phosphonate esters can serve two purposes. One [purposes] <u>purpose</u> is as a surface phosphonating agent, and the other as a surface silanating agent which will generate the hydrophobic surface necessary for incorporation into hydrophobic monomers. If the silane is alkene functionalized, or otherwise functionalized with a polymerizable functionality, then the silane also acts as a coupling agent via the phosphonating agent.

2. Page 11, paragraph encompassing line 8:

In order to make an especially active phosphonating species, the dimethyl ester of methacryl phosphonate was reacted with a silanating agent to form the hydrolytically unstable vinyl dimethyl silyl ester. The silanating agent can be a chloride, as shown below, or a bromide.

In the claims

- 1. (Amended) Functionalized [metal] <u>zirconium</u> oxide particles comprising:
- a surface comprising a total quantity of hydroxyl groups comprising a complexed fraction comprising a reactive portion and a less reactive portion;
- said reactive portion being complexed with functionalities selected from the group consisting of functionalities with high steric hindrance, functionalities with low steric hindrance, and a combination thereof;
- said less reactive portion being complexed with said groups having a low steric hindrance.
- 2. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 1 wherein said functionalities having a low steric hindrance comprise mobile adhesion promoters and said functionalities having a high steric hindrance comprise organofunctional coupling agents.
- 3. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 1 wherein said complexed fraction is effective to produce a coagulation point of about 1 minute or more.

- 4. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 1 wherein said complexed fraction is effective to produce a coagulation point of about 1 hour or more.
- 5. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 2 wherein said complexed fraction is effective to produce a coagulation point of about 1 minute or more.
- 6. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 2 wherein said complexed fraction is effective to produce a coagulation point of about 1 hour or more.
- 7. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 1 wherein said complexed fraction is about 50% or more of said total quantity.
- 8. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 2 wherein said organofunctional coupling agents are irreversibly [conplexed] <u>complexed</u> with said reactive portion.
- 9. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 1 wherein said complexed fraction comprises substantially all of said total quantity.
- 10. (Amended)The functionalized [metal] <u>zirconium</u> oxide particles of claim 2 wherein said complexed fraction comprises substantially all of said total quantity.
- 11. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 3 wherein said complexed fraction comprises substantially all of said total quantity.
- 12. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 4 wherein said complexed fraction comprises substantially all of said total quantity.

- 13. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 5 wherein said complexed fraction comprises substantially all of said total quantity.
- 14. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 6 wherein said complexed fraction comprises substantially all of said total quantity.
- 15. (Amended) Functionalized [metal] <u>zirconium</u> oxide particles comprising a surface comprising a total quantity of hydroxyl groups comprising a complexed fraction and an uncomplexed fraction, said complexed fraction being effective to produce a coagulation point of about one minute or more after removal of a solvent from a mixture of said metal oxide particles and a matrix resin.
- 16. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 15 wherein said complexed fraction is effective to produce a coagulation point of about one hour or more after removal of a solvent.
- 17. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 15 wherein said complexed portion comprises a less reactive portion complexed with a mobile adhesion promoter and a more reactive portion complexed with an organofunctional coupling agent.
- 18. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 16 wherein said complexed portion comprises a less reactive portion complexed with a mobile adhesion promoter and a more reactive portion complexed with an organofunctional coupling agent.
- 19. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 17 wherein the organofunctional coupling agent also comprises an adhesion promoter.

- 20. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 18 wherein the organofunctional coupling agent also comprises an adhesion promoter.
- 26. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 22 wherein said metal oxide comprises a metal selected from the group consisting of niobium, indium, titanium, zinc, zirconium, tin, cerium, hafnium, tantalum, tungsten, bismuth, and combinations thereof.
- 27. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 23 further comprising an alloying element selected from the group consisting of aluminum, phosphorus, gallium, germanium, barium, strontium, yttrium, niobium, antimony, cesium, and combinations thereof.
- 28. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 24 further comprising an alloying element selected from the group consisting of aluminum, phosphorus, gallium, germanium, barium, strontium, yttrium, niobium, antimony, cesium, and combinations thereof.
- 29. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 25 further comprising an alloying element selected from the group consisting of aluminum, phosphorus, gallium, germanium, barium, strontium, yttrium, niobium, antimony, cesium, and combinations thereof.
- 30. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 26 further comprising an alloying element selected from the group consisting of aluminum, phosphorus, gallium, germanium, barium, strontium, yttrium, niobium, antimony, cesium, and combinations thereof.

- 31. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 27 further comprising an alloying element selected from the group consisting of aluminum, phosphorus, gallium, germanium, barium, strontium, yttrium, niobium, antimony, cesium, and combinations thereof.
- 32. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 28 further comprising an alloying element selected from the group consisting of aluminum, phosphorus, gallium, germanium, barium, strontium, yttrium, niobium, antimony, cesium, and combinations thereof.
- 39. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 1 further comprising a mixture comprising a matrix resin comprising monomers comprising functional groups polymerizable with said organofunctional coupling agents.
- 40. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 2 further comprising a mixture comprising a matrix resin comprising monomers comprising functional groups polymerizable with said organofunctional coupling agents.
- 41. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 5 further comprising a mixture comprising a matrix resin comprising monomers comprising functional groups polymerizable with said organofunctional coupling agents.
- 42. (Amended) The functionalized [metal] <u>zircōnium</u> oxide particles of claim 6 further comprising a mixture comprising a matrix resin comprising monomers comprising functional groups polymerizable with said organofunctional coupling agents.
- 43. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 17 further comprising a mixture comprising a matrix resin comprising monomers comprising functional groups polymerizable with said organofunctional coupling agents.

- 44. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 18 further comprising a mixture comprising a matrix resin comprising monomers comprising functional groups polymerizable with said organofunctional coupling agents.
- 45. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 1 comprising an average diameter effective to permit curing of said mixture by photopolymerization.
- 46. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 2 comprising an average diameter effective to permit curing of said mixture by photopolymerization.
- 47. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 5 comprising an average diameter effective to permit curing of said mixture by photopolymerization.
- 48. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 6 comprising an average diameter effective to permit curing of said mixture by photopolymerization.
- 49. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 17 comprising an average diameter effective to permit curing of said mixture by photopolymerization.
- 50. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 18 comprising an average diameter effective to permit curing of said mixture by photopolymerization.
- 51. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 2 wherein a sufficient quantity of said reactive portion is complexed with an organofunctional

coupling agent to provide fracture toughness of a cured composite comprising said functionalized metal oxide particles.

- 52. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 5 wherein [said] a sufficient quantity of said reactive portion is complexed with said organofunctional agent to provide fracture toughness of a cured composite comprising said functionalized metal oxide particles.
- 53. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 6 wherein [said] a sufficient quantity of said reactive portion is complexed with said organofunctional agent to provide fracture toughness of a cured composite comprising said functionalized metal oxide particles.
- 54. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 17 wherein [said] a sufficient quantity of said reactive portion is complexed with said organofunctional agent to provide fracture toughness of a cured composite comprising said functionalized metal oxide particles.
- 55. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 18 wherein [said] a sufficient quantity of said reactive portion is complexed with said organofunctional agent to provide fracture toughness of a cured composite comprising said functionalized metal oxide particles.
- 56. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 2 wherein the organofunctional coupling agent comprises a polymerizable group selected from the group consisting of one or more vinyl groups, acryl groups, epoxy groups, [or] <u>and</u> methacryl groups.

- 57. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 5 wherein the organofunctional coupling agent comprises a polymerizable group selected from the group consisting of one or more vinyl groups, acryl groups, epoxy groups, [or] <u>and</u> methacryl groups.
- 58. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 6 wherein the organofunctional coupling agent comprises a polymerizable group selected from the group consisting of one or more vinyl groups, acryl groups, epoxy groups, [or] <u>and</u> methacryl groups.
- 59. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 17 wherein the organofunctional coupling agent comprises a polymerizable group selected from the group consisting of one or more vinyl groups, acryl groups, epoxy groups, [or] <u>and</u> methacryl groups.
- 60. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 18 wherein the organofunctional coupling agent comprises a polymerizable group selected from the group consisting of one or more vinyl groups, acryl groups, epoxy groups, [or] <u>and</u> methacryl groups.
- 61. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 56 wherein the organofunctional coupling agent comprises a functionality selected from the group consisting of mono-, di-, and tri-functional silanes, isocyanates, zirconates, aluminozirconates, zirconyl methacrylate, titanates, and phosphonates.
- 62. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 57 wherein the organofunctional coupling agent comprises a functionality selected from the group

consisting of mono-, di-, and tri-functional silanes, isocyanates, zirconates, aluminozirconates, zirconyl methacrylate, titanates, and phosphonates.

- 63. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 58 wherein the organofunctional coupling agent comprises a functionality selected from the group consisting of mono-, di-, and tri-functional silanes, isocyanates, zirconates, aluminozirconates, zirconyl methacrylate, titanates, and phosphonates.
- 64. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 59 wherein the organofunctional coupling agent comprises a functionality selected from the group consisting of mono-, di-, and tri-functional silanes, isocyanates, zirconates, aluminozirconates, zirconyl methacrylate, titanates, and phosphonates.
- 65. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 60 wherein the organofunctional coupling agent comprises a functionality selected from the group consisting of mono-, di-, and tri-functional silanes, isocyanates, zirconates, aluminozirconates, zirconyl methacrylate, titanates, and phosphonates.
- 66. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 2 wherein the organofunctional groups are hydrolyzable zirconates having the following general structure:

$$R^1$$
 -O- Zr - $(OR^2)_3$

wherein

- R¹ is selected from the group consisting of hydrolyzable alkyl groups and hydrolyzable alkenyl groups having 1 or more carbon atoms; and
- R² is selected from the group consisting of copolymerizable alkenyl substituents containing 2 or more carbon atoms.

67. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 5 wherein the organofunctional groups are hydrolyzable zirconates having the following general structure:

$$R^1$$
 -O- Zr - $(OR^2)_3$

wherein

R¹ is selected from the group consisting of hydrolyzable alkyl groups and hydrolyzable alkenyl groups having 1 or more carbon atoms; and

 R^2 is selected from the group consisting of copolymerizable alkenyl substituents containing 2 or more carbon atoms.

68. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 6 wherein the organofunctional groups are hydrolyzable zirconates having the following general structure:

$$R^{1}$$
 -O- Zr - $(OR^{2})_{3}$

wherein

R¹ is selected from the group consisting of hydrolyzable alkyl groups and hydrolyzable alkenyl groups having 1 or more carbon atoms; and

R² is selected from the group consisting of copolymerizable alkenyl substituents containing 2 or more carbon atoms.

69. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 17 wherein the organofunctional groups are hydrolyzable zirconates having the following general structure:

$$R^1$$
 -O- Zr - $(OR^2)_3$

wherein

- R¹ is selected from the group consisting of hydrolyzable alkyl groups and hydrolyzable alkenyl groups having 1 or more carbon atoms; and
- R² is selected from the group consisting of copolymerizable alkenyl substituents containing 2 or more carbon atoms.
- 70. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 18 wherein the organofunctional groups are hydrolyzable zirconates having the following general structure:

$$R^{1}$$
 -O- Zr - $(OR^{2})_{3}$

wherein

- R¹ is selected from the group consisting of hydrolyzable alkyl groups and hydrolyzable alkenyl groups having 1 or more carbon atoms; and
- R^2 is selected from the group consisting of copolymerizable alkenyl substituents containing 2 or more carbon atoms.
- 71. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 66 wherein R¹ is selected from the group consisting of alkyl groups having from about 1 to about 9 carbon atoms.
- 72. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 67 wherein R¹ is selected from the group consisting of alkyl groups having from about 1 to about 9 carbon atoms.
- 73. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 68 wherein R¹ is selected from the group consisting of alkyl groups having from about 1 to about 9 carbon atoms.

- 74. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 69 wherein R¹ is selected from the group consisting of alkyl groups having from about 1 to about 9 carbon atoms.
- 75. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 70 wherein R¹ is selected from the group consisting of alkyl groups having from about 1 to about 9 carbon atoms.
- 76. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 2 wherein the organofunctional groups comprise moeities selected from the group consisting of neopentyl (diallyl) oxy trimethacryl zirconates, neopentyl (diallyl) oxy triacryl zirconates, and aluminozirconates having the following general structure:

wherein R³ is selected from the group consisting of copolymerizable alkenyl groups and carboxyfunctional substituents containing 1 or more carbon atoms.

77. (Amended) The functionalized metal oxide particles of claim 5 wherein the organofunctional groups comprise moeities selected from the group consisting of neopentyl

(diallyl) oxy trimethacryl zirconates, neopentyl (diallyl) oxy triacryl zirconates, and aluminozirconates having the following general structure: wherein R³ is selected from the group consisting of copolymerizable alkenyl groups and carboxyfunctional substituents containing 1 or more carbon atoms.

78. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claims 6 wherein the organofunctional groups comprise moeities selected from the group consisting of neopentyl (diallyl) oxy trimethacryl zirconates, neopentyl (diallyl) oxy triacryl zirconates, and aluminozirconates having the following general structure:

wherein R³ is selected from the group consisting of copolymerizable alkenyl groups and carboxyfunctional substituents containing 1 or more carbon atoms.

79. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 17 wherein the organofunctional groups comprise moeities selected from the group consisting of neopentyl (diallyl) oxy trimethacryl zirconates, neopentyl (diallyl) oxy triacryl zirconates, and aluminozirconates having the following general structure:

wherein R³ is selected from the group consisting of copolymerizable alkenyl groups and carboxyfunctional substituents containing 1 or more carbon atoms.

80. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 18 wherein the organofunctional groups comprise moeities selected from the group consisting of neopentyl (diallyl) oxy trimethacryl zirconates, neopentyl (diallyl) oxy triacryl zirconates, and aluminozirconates having the following general structure:

- wherein R³ is selected from the group consisting of copolymerizable alkenyl groups and carboxyfunctional substituents containing 1 or more carbon atoms.
- 81. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 2 wherein said organofunctional groups are methacryloxy aluminozirconates.
- 82. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 5 wherein said organofunctional groups are methacryloxy aluminozirconates.
- 83. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 6 wherein said organofunctional groups are methacryloxy aluminozirconates.
- 84. (Amended) The [metal] <u>functionalized zirconium</u> metal oxide particles of claim 17 wherein said organofunctional groups are methacryloxy aluminozirconates.
- 85. (Amended) The [metal] <u>functionalized zirconium</u> oxide particles of claim 18 wherein said organofunctional groups are methacryloxy aluminozirconates.
 - 86. (Amended) Functionalized [metal] <u>zirconium</u> oxide particles comprising:
 - a surface comprising a total quantity of hydroxyl groups comprising a complexed fraction comprising a reactive portion and a less reactive portion;
 - said reactive portion being complexed with functionalities selected from the group consisting of functionalities with high steric hindrance, functionalities with low steric hindrance, and a combination thereof;
 - said less reactive portion being complexed with said groups having a low steric hindrance;
 - wherein one or more of said functionalities with high steric hindrance and said functionalities with low steric hindrance is bound to the oxide surface via an ester linkage to a phosphonate group.

- 87. (Amended) The [method] <u>functionalized zirconium oxide particles</u> of claim 2 wherein one or more of said organofunctional coupling agent and said mobile adhesion promoter is bound to the oxide surface via an ester linkage to a phosphonate group.
- 88 (Amended) The [method] <u>functionalized zirconium oxide particles</u> of claim 5 wherein one or more of said organofunctional coupling agents and said mobile adhesion promoter is bound to the oxide surface via an ester linkage to a phosphonate group.
- 89. (Amended) The [method] <u>functionalized zirconium oxide particles</u> of claim 6 wherein one or more of said organofunctional coupling agents and said less reactive functionalities is bound to the oxide surface via an ester linkage to a phosphonate group.
- 90. (Amended) The [method] <u>functionalized zirconium oxide particles</u> of claim 17 wherein one or more of said organofunctional coupling agents and said less reactive functionalities is bound to the oxide surface via an ester linkage to a phosphonate group.
- 91. (Amended) The [method] <u>functionalized zirconium oxide particles</u> of claim 18 wherein one or more of said organofunctional coupling agents and said less reactive functionalities is bound to the oxide surface via an ester linkage to a phosphonate group.
- 92. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 86 wherein said phosphonate group comprises a silyl ester which may or may not comprise a polymerizable group.
- 93. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 87 wherein said phosphonate group comprises a silyl ester which may or may not comprise a polymerizable group.

- 94. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 88 wherein said phosphonate group comprises a silyl ester which may or may not comprise a polymerizable group.
- 95. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 89 wherein said phosphonate group comprises a silyl ester which may or may not comprise a polymerizable group.
- 96. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 90 wherein the phosphonate group comprises a silyl ester which may or may not comprise a polymerizable group.
- 97. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 91 wherein the phosphonate group comprises a silyl ester which may or may not comprise a polymerizable group.
- 98. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 1 wherein the functionality with low steric hindrance is selected from the group consisting of silanes, phosphonates, phosphates, chelating agents, fatty acids, fatty alcohols, and ester linked fatty acids.
- 99. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 2 wherein the mobile adhesion promoter is selected from the group consisting of silanes, phosphonates, phosphates, chelating agents, fatty acids, fatty alcohols, and ester linked fatty acids.
- 100. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 5 wherein the mobile adhesion promoter is selected from the group consisting of silanes,

phosphonates, phosphates, chelating agents, fatty acids, fatty alcohols, and ester linked fatty acids.

- 101. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 6 wherein the mobile adhesion promoter is selected from the group consisting of silanes, phosphonates, phosphates, chelating agents, fatty acids, fatty alcohols, and ester linked fatty acids.
- 102. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 17 wherein the mobile adhesion promoter is selected from the group consisting of silanes, phosphonates, phosphates, chelating agents, fatty acids, fatty alcohols, and ester linked fatty acids.
- 103. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 18 wherein the mobile adhesion promoter is selected from the group consisting of silanes, phosphonates, phosphates, chelating agents, fatty acids, fatty alcohols, and ester linked fatty acids.
- 104. (Amended) The [metal] <u>functionalized zirconium</u> oxide particles of claim 2 wherein the mobile adhesion promoter comprises dimethyl ethoxy vinyl silane.
- 105. (Amended) The [metal] <u>functionalized zirconium</u> oxide particles of claim 5 wherein the mobile adhesion promoter comprises dimethyl ethoxy vinyl silane.
- 106. (Amended) The [metal] <u>zirconium</u> oxide particles of claim 6 wherein the mobile adhesion promoter comprises dimethyl ethoxy vinyl silane.
- 107. (Amended) The [metal] <u>zirconium</u> oxide particles of claim 17 wherein the mobile adhesion promoter comprises dimethyl ethoxy vinyl silane.

- 108. (Amended) The [metal] <u>zirconium</u> oxide particles of claim 18 wherein the mobile adhesion promoter comprises dimethyl ethoxy vinyl silane.
- 109. (Amended) The [metal] <u>zirconium</u> oxide particles of claim 1 having an average diameter of from about 10 to about 150 nanometers.
- 110. (Amended) The [metal] <u>zirconium</u> oxide particles of claim 2 having an average diameter of from about 10 to about 150 nanometers.
- 111. (Amended) The [metal] <u>zirconium</u> oxide particles of claim 3 having an average diameter of from about 10 to about 150 nanometers.
- 112. (Amended) The [metal] <u>zirconium</u> oxide particles of claim 4 having an average diameter of from about 10 to about 150 nanometers.
- 113. (Amended) A composition comprising the [metal] <u>zirconium</u> oxide particles of claim 2 and a matrix comprising at least one monomer comprising a group polymerizable with the organofunctional coupling agent.
- 114. (Amended) A composition comprising the [metal] <u>zirconium</u> oxide particles of claim 5 and a matrix comprising at least one monomer comprising a group polymerizable with the organofunctional coupling agent.
- 115. (Amended) A composition comprising the [metal] <u>zirconium</u> oxide particles of claim 6 and a matrix comprising at least one monomer comprising a group polymerizable with the organofunctional coupling agent.
- 116. (Amended) A composition comprising the [metal] <u>zirconium</u> oxide particles of claim 18 and a matrix comprising at least one monomer comprising a group polymerizable with the organofunctional coupling agent.

- 117. (Amended) A composite comprising the composition of claim 113 wherein said group and said organofunctional coupling agent are copolymerized.
- 118. (Amended) A composite comprising the composition of claim 114 wherein said group and said organofunctional coupling agent are copolymerized.
- 119. (Amended) A composite comprising the composition of claim 115 wherein said group and said organofunctional coupling agent are copolymerized.
- 120. (Amended) A composite comprising the composition of claim 116 wherein said group and said organofunctional coupling agent are copolymerized.
- 121. (Amended) The composition of claim 113 comprising a dental restorative composition.
- 122. (Amended) The composition of claim 117 comprising a dental restorative composition.
 - 123. (Amended) The composition of claim 113 comprising a prototyping composition.
 - 124. (Amended) The composition of claim 117 comprising a prototyping composition.
- 128. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 1 wherein the organofunctional groups comprise moeities selected from the group consisting of neopentyl (diallyl) oxy trimethyacryl zirconates and neopentyl (diallyl) oxy triacryl zirconates.
- 129. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 2 wherein the organofunctional groups comprise moeities selected from the group consisting of neopentyl (diallyl) oxy trimethyacryl zirconates and neopentyl (diallyl) oxy triacryl zirconates.
- 130. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 66 wherein the organofunctional groups comprise moeities selected from the group consisting of neopentyl (diallyl) oxy trimethyacryl zirconates and neopentyl (diallyl) oxy triacryl zirconates.

131. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 70 wherein the organofunctional groups comprise moeities selected from the group consisting of neopentyl (diallyl) oxy trimethyacryl zirconates and neopentyl (diallyl) oxy triacryl zirconates.

132. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 71 wherein the organofunctional groups comprise moeities selected from the group consisting of neopentyl (diallyl) oxy trimethyacryl zirconates and neopentyl (diallyl) oxy triacryl zirconates.

133. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim 75 wherein the organofunctional groups comprise moeities selected from the group consisting of neopentyl (diallyl) oxy trimethyacryl zirconates and neopentyl (diallyl) oxy triacryl zirconates.

134. (Amended) The functionalized [metal] <u>zirconium</u> oxide particles of claim [2] <u>3</u> wherein the mobile adhesion promoter is selected from the group consisting of silanes, phosphonates, phosphonates, chelating agents, fatty acids, fatty alcohols, and ester linked fatty acids.

Respectfully submitted.

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